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3ω Damage: Growth Mitigation

M. Kozlowski, S. Demos, Z-L Wu, J. Wong, B. Penetrante, L. Hrubesh

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LDRD FINAL REPORT

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Introduction and purpose

The design of high power UV laser systems is limited to a large extent by the laser-initiated damage performance of transmissive fused silica optical components. The 3ω (ie., the third harmonic of the primary laser frequency) damage growth mitigation LDRD effort focused on understanding and reducing the rapid growth of laser-initiated surface damage on fused silica optics. Laser-initiated damage can be discussed in terms of two key issues: damage initiated at some type of precursor and rapid damage growth of the damage due to subsequent laser pulses. The objective of the LDRD effort has been the elucidation of laser-induced damage processes in order to quantify and potentially reduce the risk of damage to fused silica surfaces. The emphasis of the first two years of this effort was the characterization and reduction of damage initiation. In spite of significant reductions in the density of damage sites on polished surfaces, statistically some amount of damage initiation should always be expected. The early effort therefore emphasized the development of testing techniques that quantified the statistical nature of damage initiation on optical surfaces. This work led to the development of an optics lifetime modeling strategy that has been adopted by the NIF project to address damage-risk issues [1,2]. During FY99 interest shifted to the damage growth issue which was the focus of the final year of this project. The impact of the remaining damage sites on laser performance can be minimized if the damage sites did not continue to grow following subsequent illumination. The objectives of the final year of the LDRD effort were to apply a suite of state-of-the-art characterization tools to elucidate the nature of the initiated damage sites, and to identify a method that effectively mitigates further damage growth. Our specific goal is to understand the cause for the rapid growth of damage sites so that we can develop and apply an effective means to mitigate it.

The prevailing hypothesis for the growth mechanism of laser-initiated damage involves a synergism of some means for absorption of 3ω light at the damage site and local field enhancement due to cracks. A proposed mechanism for damage growth involves an initial damage at a precursor resulting in the transformation of basically non-absorbing SiO_2 to form an absorbing layer of d-SiOx. In this context d-SiOx implies SiO_2 modified in terms of either the formation of other stoichiometries (eg., SiO, Si, or more generally SiOx with $0<x<2$) or significant concentrations of defects (broken bonds, vacancies, etc.). Earlier efforts focused on the characterization of the absorption mechanisms and measurement of the field enhancement due to cracks. The FY00 effort continued the identification of the absorbing species and the characterization of damage morphology while emphasizing the development of growth mitigation techniques directed at removing

both the absorbing species and the cracks. We applied a variety of analytical tools to characterize the damage morphology, including; photoluminescence (PL) spectroscopy, optical and photothermal microscopies, high resolution transmission electron microscopy (TEM) and electron-spin-resonance (ESR) spectroscopy, x-ray photoelectron spectroscopy (XPS) , secondary ion mass spectrometry (SIMS), x-ray micro-tomography (XMT) and cathodo-luminescence (CL).

The objective of the surface damage mitigation effort is to experimentally validate methods that could effectively stop the growth of 3ω laser-initiated damage. A specific goal is to obtain data and information on successful methods for fused silica optics, which would be sufficient to enable the down-selection to a single approach. Future effort could then be focused on developing a primary method for actual implementation on NIF. It is also the intent of this study to prioritize the remaining successful methods, so that there will be a back-up selection if the primary method fails to meet requirements. The mitigation methods selected for the study are chemical etching, plasma-etching, CO_2 laser processing, and micro-flame torch processing. The number of experiments differed for the various methods and therefore the data for some techniques is limited. Nevertheless, there is sufficient data and information to choose a primary mitigation method. The CO_2 laser processing shows the most significant effect to halt damage growth, and there is already enough data to have confidence in the approach for mitigating damage on NIF relevant silica optics.

Major accomplishments and results for FY98 - FY99

1) Surface damage initiation: Experimental and modeling efforts indicated that the remaining precursors for fused silica laser damage are 10nm-scale absorbing defects [3]. While significant progress has been made in the development of near-field optical microscopy (NSOM) and photothermal (PT) imaging [4] as precursor identification tools, it is unlikely that all precursors will be eliminated. Our efforts have therefore emphasized the development of measurement techniques and a data analysis model based on Weibull statistics to quantify the risk of damage to large-aperture high-quality optics. Using data measured in off-line facilities, the model successfully predicted the density of damage sites produced on a lens during a Beamlet frequency conversion campaign [2]. Activities associated with measurement of precursor densities and the refinement of optic lifetime models were transferred to the NIF Project.

2) Surface damage growth: A mechanism based on the damage-induced formation of absorbing d-SiOx layers was proposed. In the course of damage studies it was observed that damage sites on fused silica luminesce when exposed to UV light. We investigated the luminescence signals as a path to identifying the d-SiOx species. Luminescence studies were performed using 351 nm CW excitation of damage sites created using 355 nm, 7.5 ns laser pulses. The experimental results reveal the presence of optically active defects within the laser damage sites. The spectra in Fig. 1 shows at least three characteristic luminescence bands: 1.9 eV (650 nm), 2.2 eV (560 nm) ~4.7 eV

(~440 nm). The 650 nm and 440 nm bands are likely associated with the non-bridging oxygen center (NBOHC)(ie., broken Si-O bonds) and the oxygen vacancy center (E'), respectively [5,6]. The presence of the NBOHC is supported by the observation of a characteristic Raman peak at 890 cm^{-1}. The band at 560 nm has not been positively identified although it is likely associated with a self-trapped exciton state or with silicon nanoparticles [7]. The emission spectrum of the damaged material is non-uniform across the damage site indicating a non-uniform composition of the d-SiOx layer. Photothermal (PT) mapping studies have also been performed on the damage sites to more directly detect local laser induced-heating. Non-uniform heating within the damage site was observed with features similar to those in the luminescence maps. The luminescence and photothermal data are consistent with the proposed "absorbing d-SiOx" model for laser damage growth.

We have investigated chemical etching of the absorbing layer as a possible mitigation technique for continued damage growth. Figure 2 shows that the intensity of emission was reduced by chemical etching with a HF/HNO_3 solution. The difference in the post-etch spectra for the 2- and 5-pulse sites indicates that the thickness/composition of the d-SiOx layer is dependent on the illumination history of the damage site. We began etching studies to examine the dependence of the d-SiOx layer thickness/composition on the number and fluence of the laser damage pulses. The remainder of FY99 focused on damage growth rate studies to demonstrate that the reduction in luminescence can be correlated with the decrease in the growth rate.

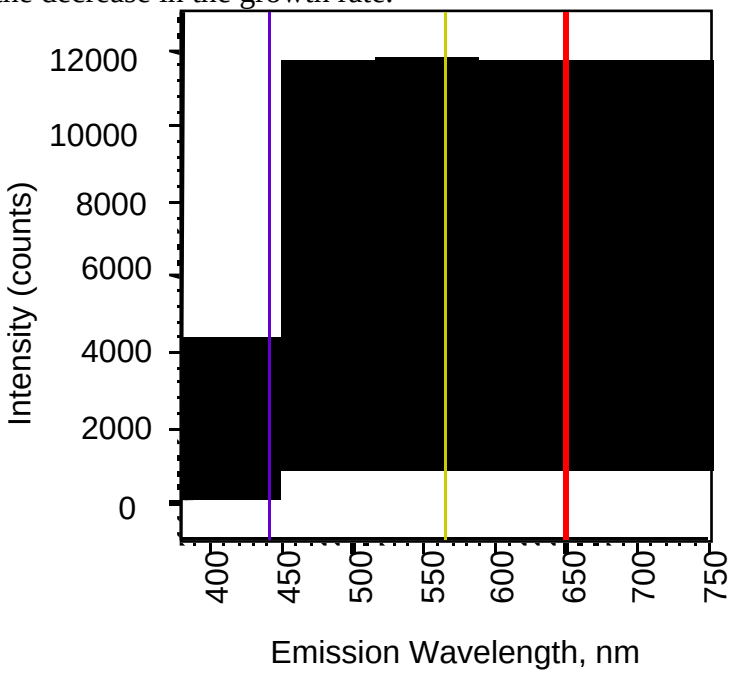

Figure 1: Emission spectra of damage created with one 40 J/cm^2 pulse from a 355 nm laser. The spectrum is obtained from an area with diameter of 5 μm. The primary emission peaks at 440 nm, 560 nm and 650 nm are indicated.

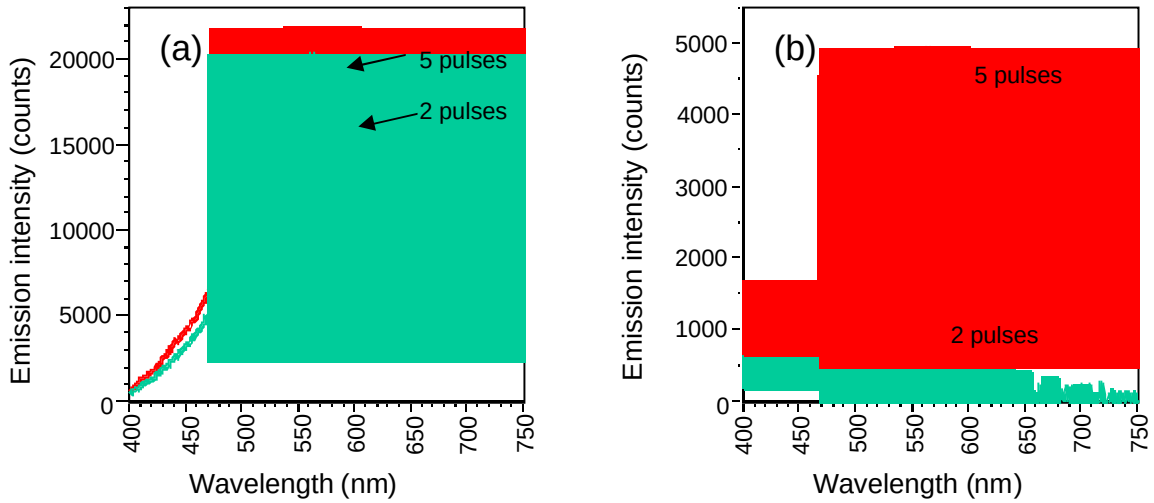

Figure 2: Emission spectra measured for damage sites created with 2 pulses and 5 pulses at 40 J/cm^2, 355nm, 7.5 ns, (a) before and (b) after 11 minutes (~1.1 µm) of etch with the HF/HNO_3 solution. Note the change in the y-axis scale.

Major accomplishments and results for FY00

A. Damage characterization studies

1) Damage morphology: The morphology of damaged regions in fused silica which result from irradiation of a pulse of high fluence 3ω light, consists of a group of pits spanning a fraction of the $1/e^2$ beam size incident on the sample. Figure 3 shows such a typical damage morphology induced by a pulse of ~45 J/cm^2, 3ω light having a temporal pulse width of 7.6 ns and a $1/e^2$ beam diameter of 0.98 mm. The spatial profile of the 3ω beam is very nearly Gaussian.

Systematic SEM examinations show that the damage sites consist primarily of a once molten core region (possibly produced by thermal explosion), surrounded by a near concentric region of fractured material, indicative of mechanical damage accompanied by spallation of material (see Fig. 3(b)). The latter arises from propagation of lateral cracks induced by the laser-generated shock waves. The size of the overall crater depends on laser fluence, the number of pulses, the laser irradiation history and the test environment. A compacted layer, ~10 microns thick and ~20% higher density than the bulk, has been identified with XMT, at the bottom of the damage crater. We mapped regions of high absorption within the damage sites and we showed that the densified layer within the damage sites contain electron paramagnetic (E' centers), non-bonding oxygen hole centers (NBOHC) and oxygen deficient centers (ODC (II)). High-resolution microprobe analysis shows that there is no variation in the Si/O stoichiometry in the compacted layer, to within ±1.6%. High resolution TEM indicates the absence of crystalline nano-particles of Si in the compacted layer. Four point defects: the E'_{γ}, E'_{74}, NBOHC, and ODC (II), have been identified using a combination of ESR and CL micro-spectroscopy. We were able to correlate the PL spectral signatures as well as the CL results, with certain defects in silica such as the NBOHC and ODC (II) defects.

Stereographic SEM imaging reveals the depth morphology of the damage pits. The depth profile along a selected cross section may be obtained quantitatively by simple geometry using $z = P/[2M(\sin\alpha/2)]$, where z is the depth, P the parallax, M magnification of the SEM measurement and α is the tilt angle between the stereo pair of images. The results are shown in Fig.4. Combined with white-light interferometry, the aspect ratio of these high fluence damage pits was determined to be 0.24 ± 0.05. This ratio is similar to those observed and calculated for craters created by both meteor impacts as well as by underground explosions [8].

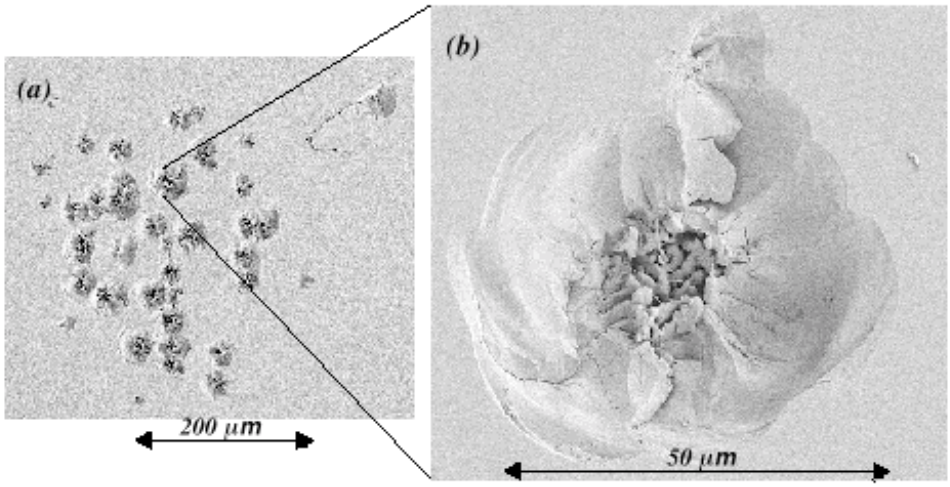

Figure 3(a) SEM micrograph of a damage site in fused silica induced by 1 pulse of 3ω light at 45J/cm ^{2}, showing a cluster of pits; (b) a single damage crater revealing a central melted core with a fractured annulus.

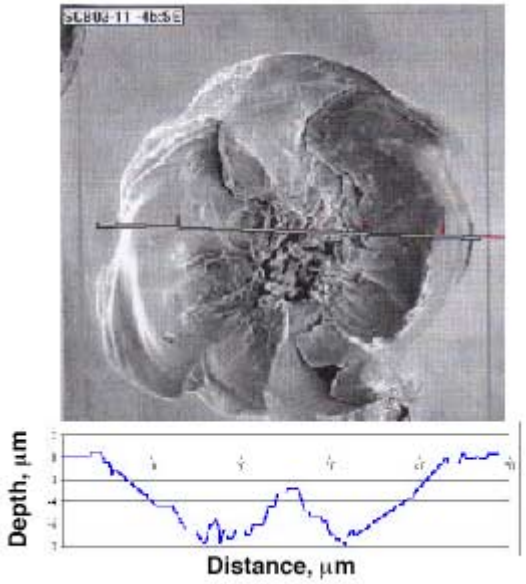

Figure 4. Stereo SEM anaglyph of the damage crater shown in Fig. 3(b) used for depth profiling (bottom plot).

2) Si/O stoichiometry and water content : The Si/O stoichiometry in the region of the damage pit has been determined with electron microprobe analysis. The Si/O ratio at the bottom of the damage pits to a depth of 100 micrometers was found invariant with that of an undamaged fused silica standard, within experimental accuracy. The base fused silica material is Corning 7980 which contains 800-1000 ppm of "water". SIMS profile analysis on a typical damage pit revealed that there is discernable hydrogen loss from the damage region when normalized to the hydrogen content of the undamaged area of the same optic. The results are plotted in Fig. 5.

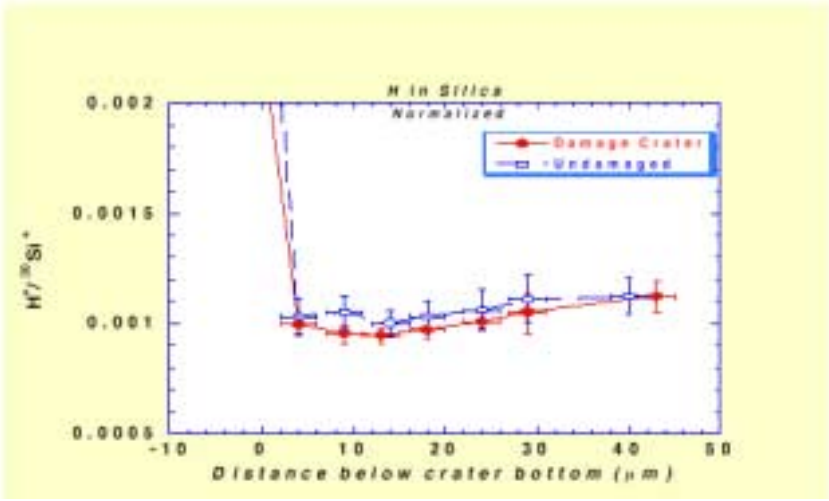

Figure 5. SIMS depth profile analysis for H loss immediately below the damage crater (shown in Fig.3b) compared with undamaged area in the same fused silica optic.

3) Densification within the crater: At a given chemical composition, the x-ray absorption value of a material is a direct measure of its physical density as evident from Beer's Law, since x-ray attenuation in matter is a mass absorption effect. XMT was applied to detect densification in the region of the 3ω damage pits. Figure 6(a) is a tomographic slice, 3.3μm thick, rendered with the volume absorption data and it reveals a cross sectional x-ray radiographic image of the damage site. The absorption profile in Fig. 6(b) shows a ~20% higher absorption value than the constant bulk value, at the bottom of the crater to a depth of ~10 μm. Figure 7 shows a 3-dimensional rendering of the compacted layer. The compact layer is not quite continuous at the wall of the pit in the vicinity of the fractured annulus region.

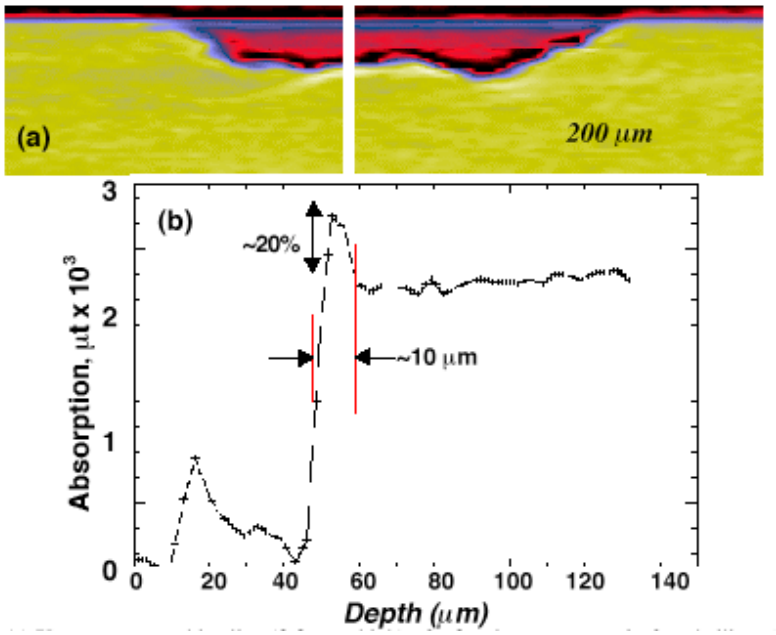

Figure 6. (a) X-ray tomographic slice (3.3 μm thick) of a 3ω damage crater in fused silica; (b) x-ray absorption profile along a vertical traverse indicated in (a), showing a higher density layer of ~10 μm thick at the bottom of the crater.

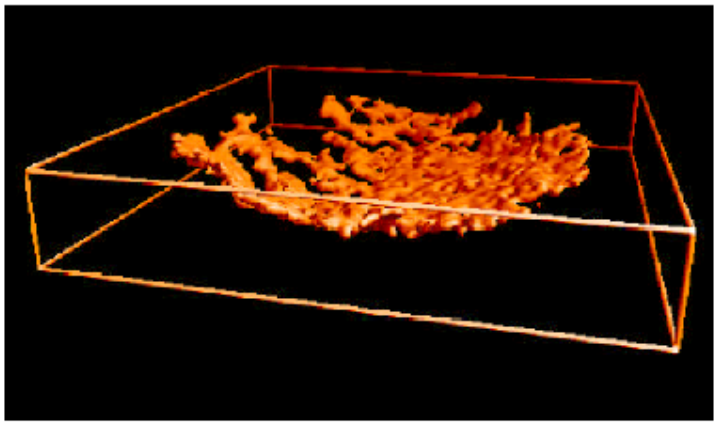

Figure 7. A 3-dimensional x-ray tomographic rendering of the compaction layer at the bottom of a 3ω damage crater in fused silica shown in Fig. 6.

4) Defects and their spatial distribution: It is well documented that a host of point defects can be generated in the fused silica structure upon irradiation with energetic particles such as neutrons, ions and electrons, and with high-energy electromagnetic radiation such as hard x-rays and gamma rays [9]. These defects may be electronic in nature, involving

a re-distribution of the local electron density to give rise to paramagnetic (eg., E'), as well as diamagnetic centers. Or the defects may be structural in nature, involving local atomic displacements from the normal random network structure of fused silica. The latter defects may be oxygen-deficient or oxygen-excess. These radiation-induced defects have been extensively studied and characterized over the last few decades using a variety of spectroscopic probes [10].

In our study, we utilized a combination of CL micro-spectroscopy and high sensitivity ESR spectroscopy, to detect the point defects in fused silica initiated by 3ω laser pulses. We were successful in identifying the E'_{γ}, , E'_{7} , NBOHC and ODC(II) centers. The ESR spectrum of the E'_{γ} center is shown in Fig. 8 , which also reveals a doublet spectral feature about the E'_{γ} signal. This doublet arises from the hyperfine interaction as a result of hydrogen substitution of one of the bridging oxygens in the E' center, hence the name E'_{74} center. The concentration of the E'_{γ} defect was determined to be 5 $\pm$ 0.5e^{11}/pulse and that for the E'_{74} was ~1.8e^{11}/pulse.

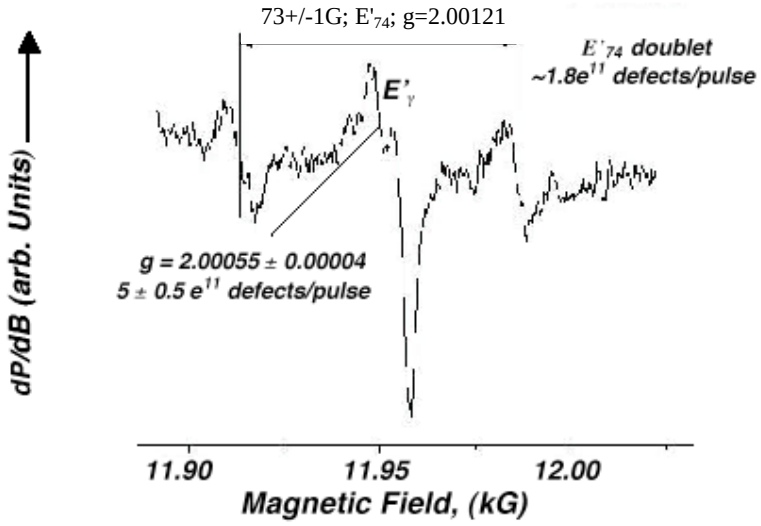

Figure 8. ESR spectrum showing existence of E'_{γ} and E'_{74} centers in high fluence (30J/cm^2, 7.5 ns, 1/e^2 diameter = 0.9 mm) 3ω damage fused silica sites. Spectrum was recorded at the Q-band (33.5 GHz) and at 4.3 K.

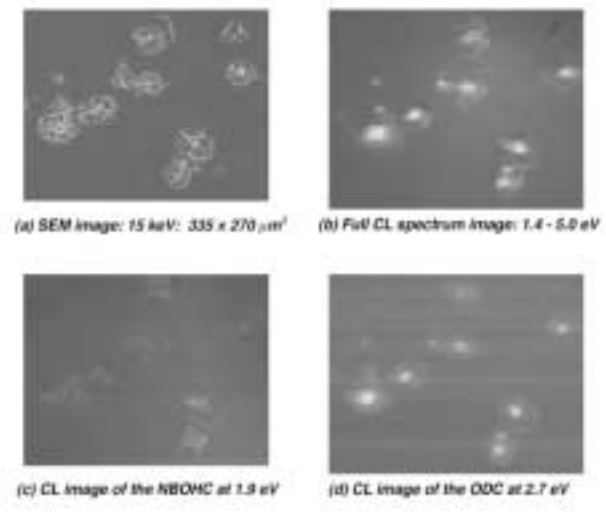

Figure 9. CL micro-spectroscopic imaging of 3ω damage craters in fused silica, showing the existence of NBOHC (1.9 eV) and ODCII (2.7 eV) and their spatial distribution. The damage was produced with 1 pulse of 3ω light at 40J/cm ^{2} , 7.6 ns and 1/e ^{2} diameter = 0.9mm

Figure 9 shows the CL data for the NBOHC and ODC (II) defects and their spatial distribution, using monochromatic imaging. It can be seen that the NBOHC defect is rather de-localized around the damage pit (Fig. 9 c) whereas the ODC (II) defect is spatially concentrated at the center of damage pits (Fig. 9d) associated with the molten nodules shown the SEM micrograph, Fig. 3b. Depth profilies of the defects were obtained by varying the energy of the electron beam in the CL microscope. Preliminary data fitting shows that the maximum concentration of defects resulting from the laser induced damage is found at the surface, consistent with damage initiation at the surface. The concentration of laser-initiated damage defects continuously decreases within the specimen, extending to a maximum depth of ~10μm. The majority of the defects are found within the first ~5μm.

B. Mitigation studies

1) Experimental procedures: In our experimental program, we tested both the intrinsic growth behavior of the mitigation pits themselves and the growth of mitigated laser-initiated damage pits. It was necessary to determine that the mitigation methods themselves did not cause damage to the fused silica surface, which would grow with repeated illumination. In fact, this was the case for those methods that tended to contaminate the surface of the fused silica. All of the experiments used the same sample type, Corning 7980 fused silica, polished by SESO (a French company), 50mm diameter, 10mm thick. For tests of bare mitigation pits, the pits were applied to one surface in an array of 6-9 spots spaced by 10mm. For the laser-initiated pits, we chose a common set of experimental conditions to place an array of 4,6, or 9, nominally uniform, equally spaced damage pits on the output surface of fused silica samples. Each laser-initiated damage site was produced by a single pulse from a laser at 355nm, with a 0.9mm beam diameter, a fluence of ~45J/cm^2 and a pulse length ~7.5ns. The typical surface damage produced by a single pulse at ~45J/cm^2 consists of a cluster of 3 to 15 pits within about 0.3mm diameter; each pit in the group has a diameter in the range from 10-50μm (see Fig. 3).

The growth tests were carried out in a vacuum chamber, in the slab laser facility. The sites were tested for growth using a tripled slab Nd:YAG laser, producing a 4 x 6mm beam at 355 nm, using ~10ns pulses at 6-12 J/cm^2. The rear surface damage sites are tested for growth. The sites are illuminated by the laser at a rate of 1 pulse per 2 seconds. If the site grows, the growth rate is determined by measuring the occluded area as a function of the number of laser shots on a site. From earlier LLNL work [11], it is known that above a threshold 3ω fluence of about 5J/cm^2, the typical damage area grows at an exponential rate with the number of laser shots. In some cases, mitigated damage had a higher threshold for growth or the growth rate was not typical. Such results were duly noted, but the growth behavior itself was not explored with further testing.

The following sections describe the experiments and results of damage growth tests for the four mitigation methods considered in this study.

2) Chemical etching experiments and results: Chemical etching with a hydrofluoric acid solution is a common method for dissolving silica and it is an accepted way to remove damage affected material on silica surfaces. Ideally, this process can return the damage area to its original state. This method was explored in previous work at LLNL to determine if mild etching (ie., to a depth of ~600nm) would remove precursors to laser-initiated damage [12,13]. The results showed a minor improvement over non-etched surfaces.

Since some change in the growth threshold was observed in earlier studies, it was reasonable to explore much deeper etching for growth mitigation because the laser-initiated damage can extend several microns deep into the material. We used 2% HF solutions to etch the entire surface of some damaged silica samples and we applied the solution directly to the damage pit, in other cases. Etch depths ranged from 0.5μm to 20μm. Growth tests were performed in slab laser facility. A summary of the results of mitigation by etching at different etch depths is shown in table 1.

Table 1. Tabulated results for growth tests for sites treated with a 2% HF solution etch.

Sample	Site	Etch depth(μm)	Test fluence (J/cm^2)	Shots to growth
SC40021	G	10	7.8	1000+
	D	10	6.8	1000+
	I	10	8.9	1000+
	F	10	9.4	40
	H	10	8.4	1000+
	E	10	8.4	1000+
SC40023	C	0.5	7.8	3
	A	1	7.5	1
	I	3	8.4	2
	G	5	8.1	3
	E	10	8.1	1
SC40026	A	10	8.2	24
	B	10	8.1	17
	C	10	8.1	830
	D	10	7.8	1000+
	E	10	8.5	19
	F	10	8.4	23
SC40027	F	20	8.4	10
	E	20	8.2	1000+
	H	20	8.5	3
	G	20	7.8	1000+

The results of our measurements show that damage sites that have been etched to depths greater than about 9 μm have about a 40% chance for zero growth with 1000 shots at fluences of 6.8-9.4 J/cm^2. For the etched sites that grow at this fluence, the growth rates are consistent with those for non-etched sites. Figure 10 shows the growth data for samples that were etched to depths of 10μm and 20μm, as they compare to the statistical

range of data for un-etched damage sites. These results are encouraging for possible mitigation of surface damage. This work was presented at the 32nd Symposium on Optical Material for High Power Lasers, and it was published in the Proceedings [14]. More data is needed for smaller damage sites to determine if the statistics for complete mitigation of growth improve for such sites.

3) Plasma-etching experiments and results: A radio-frequency (RF) plasma, spiked with fluorine-containing molecules, is commonly used to etch silica for integrated circuit applications. Fluorine atoms generated in the plasma, chemically attack the silicon-oxygen bonds of silica, forming a gas (SiF_4) which etches the material. Two types of plasma torches were investigated. One type was a miniature version of a RF argon plasma torch (micro-plasma), fed by carbon tetrafluoride (CF_4) gas, which could etch small sites on fused silica. The plasma diameter is about 1.5mm at the tip.

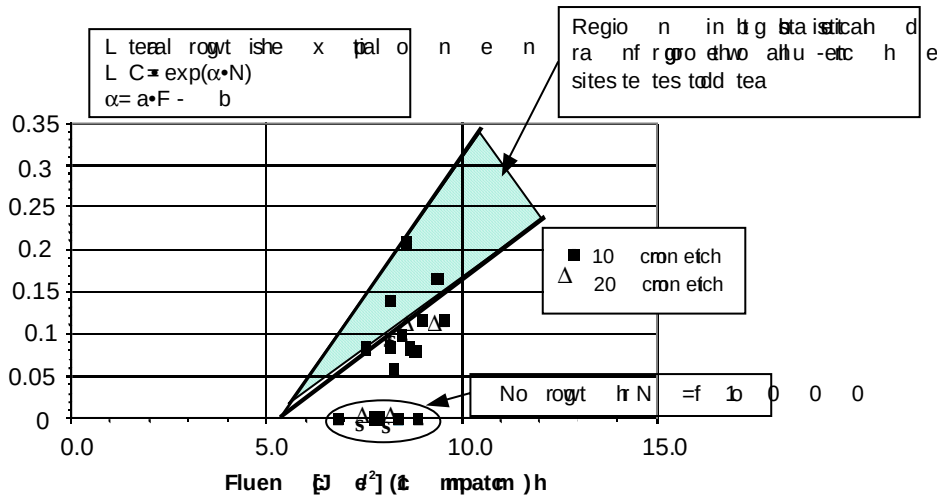

Figure 10. Plot showing results of growth experiments for wet chemical etched damage sites relative to the range of data for un-etched sites. About 40% of the etched sites did not grow for 1000 shots at the given fluences.

When the torch is applied directly to the surface of the silica, it produces a pit ~2mm diameter and the depth (~1-5µm) depends on the exposure time. A typical pit profile is shown in Fig. 11. The second type of torch was a microwave plasma torch (nitrogen gas) also fed by CF_4 gas, to produce fluorine atoms for reactive atom plasma processing (RAPP). The large RAPP torch etched silica at a fast rate (~0.2µm/min) over a diameter of 10mm. The depth (~10-100µm) depends on the exposure time.

The initial work with the micro-plasma torch exposed several problems; 1) the etch rate is very slow (~2µm/hr), 2) the etching does not rapidly remove the damage pit, and 3) the plasma produces contamination residue in the treated area. Evidence for 2) is seen in the profile shown in **Fig. 11**, where the original damage pit has receded into the substrate without smoothing as the etching takes place. Apparently, etching by the micro-torch is not isotropic as is typically the case for chemical solution etching. These sites were not

tested for growth because there was not a significant change in the damage structure after etching.

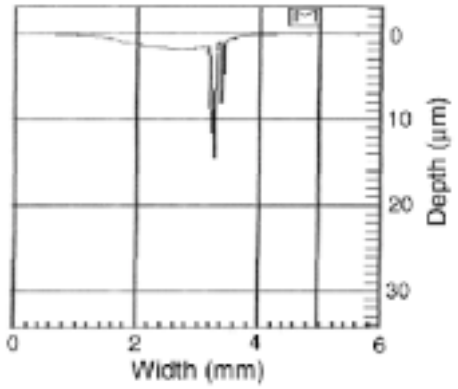

Figure 11. Profile of a 2µm deep pit micro-plasma etched over a 10-15µm laser-initiated damage site in fused silica.

Microscopy of the pits produced by the plasma torch processes before the tests, disclosed some deposits located within each pit. While the composition of the deposits have not been verifed by analysis, they are believed to be carbon residue from dissociation of the CF_4 in the plasma. The laser fluence for testing the growth in the slab laser facility was 6-8J/cm^2. All of the tested sites developed a "stain" within the first 20 shots, as they were illuminated. These stains spread in the next 20-50 laser shots, to totally fill the original plasma-produced pit. The stains appeared to originate generally at or near visible contamination within the pits.

The results of tests for both of the plasma torch processes, show that contamination in the mitigation pit tended to grow at the test fluences. These processes need additional development using non-carbon fluorine containing compounds. Also, development is needed to speed the slow etch rate of the micro-plasma torch. Due to the failure of the bare mitigation pits to survive laser illumination without contamination growth, we did not proceed to test either of the plasma torch methods with mitigated damage sites.

4) CO_2 laser experiments and results: CO_2 lasers have been used successfully by others for mitigating 1ω damage on fused silica [15,16]. We therefore explored using CO_2 lasers for mitigating 3ω damage on silica. For these tests we employed an available laser that is used for cutting, welding, and research, in the machine shop. The ~1KW, CW CO_2 laser produced pits in a silica substrate with 1 second exposure. The pits were very smooth and it was found that the pit depth could be controlled with laser power and beam focus. An example of the pit profile is shown in Fig. 12. The first growth tests in slab laser facility showed that the CO_2 pits produced on bare, undamaged silica, did not grow at fluences up to ~8J/cm^2. The actual mitigation tests included 17 damage sites that were processed with the CO_2 laser under different settings of power and focus. A summary of the tests that were done for CO_2 mitigation by melting, is shown in table 2. One site (SC40031-B)

that was shot multiple times during laser damage initiation, which had very deep cracks associated with it, grew immediately. All of the other 16 sites tested in the range of 6.7 to 12J/cm^2, 11ns, did not grow in 1000 or more shots, exhibiting complete mitigation of damage growth.

With the initial success of CO_2 laser processing to mitigate damage growth, two issues were identified for further consideration; 1) does the mitigated pit geometry affect the wavefront propagation which could damage adjacent optics, and 2) would simple thermal annealing by the CO_2 laser be sufficient to mitigate damage growth? A modeling effort was applied to the wavefront question. While the effort is not completed at this time, it appears that the affect is minimized if the diameter of the pit is less than 200µm. It should be possible to achieve this size if the laser damage itself is small enough. An experiment was done to test whether simple annealing by the CO_2 laser was sufficient to eliminate

Figure 12. An image and a profile of a pit produced on the surface of fused silica by a 1 second, 75W pulse from a CO_2 laser.

damage growth. Four sites on a 50cm dia. sample were treated by the CO_2 laser with conditions that were not sufficient to cause melt of the damage affected region (17.5W at 2.5cm defocus, for 60 sec.). The four sites were tested in slab laser facility, and all sites grew rapidly at 8J/cm^2, 11ns. These results are included in the table 2 (sample SC40031-E,G,H,I).

Table 2. Tabulated results for growth tests of sites treated with a CO_2 laser.

Sample	Site	CO_2 treatment	# Shots @ Test fluence (J/cm^2)	Comments
SC40036	A	50W,1sec	1000 @ ~6.7	no growth
	B	50W,1sec	1000 @ ~6.7	no growth
	C	none	35 @ ~6.7	control, normal growth
	D	50W,1sec+µplasma	1000 @ ~6.7	darken, no growth
SC40037	A	37.5W, 1sec	1000 @ ~8.0	no growth

	B	37.5W, 1sec	1000 @ ~8.0	no growth
	C	37.5W, 1sec	1000 @ ~8.0	no growth
SC40029	I	37.5W, 1sec+µplasma	1000 @ ~8.3	darken, no growth
	H	37.5W, 1sec	1000 @ ~8.3	no growth
	G	37.5W, 1sec+µplasma	1000 @ ~8.3	darken, no growth
	D	none	20 @ ~8.3	control, normal growth
	C	37.5W, 1sec+µplasma	1000 @ ~8.3	dnstrm optics damage
	B	37.5W, 1sec	1000 @ ~8.3	no growth
	A	37.5W, 1sec+µplasma	400 @ ~8.3	dnstrm optics damage
			1000 @ ~12.0	darken, no growth
	H	37.5W, 1sec	1000 @ ~12.0	no growth
SC40031	E	17.7W, 60sec	4 @ ~12.0	grew on first shot
	D	27.5W, 1sec	1000 @ ~12.0	no growth
	F	none	6 @ ~12.0	control, grew first shot
	A	27.5W, 1sec	1000 @ ~12.0	no growth
	B	27.5W, 1sec	7 @ ~12.0	xtra heavy damage
	C	27.5W, 1sec	1000 @ ~11.0	no growth
	I	17.5W, 60sec	10 @ ~10.0	grew first shot
	H	17.5W, 60sec	5 @ ~12.0	grew first shot
	G	17.5W, 60sec	11 @ ~11.0	grew first shot

The results support the conclusion that CO_2 laser treatment clearly demonstrates an ability to mitigate the growth of heavy (~0.3mm dia.) damage in fused silica. The process is not complex and it is amenable to rapid processing of a large number of sites on a given optic. It may also be possible to achieve growth mitigation by treating the entire optic surface, rather than point-by-point; this would eliminate the difficult, tedious task to locate specific damage sites.

5) Micro-flame torch experiments and results: We tested another method of producing local melting of the laser-damage area using a miniature version of a hydrogen flame torch. The torch was fed by CF_4 gas to produce a hot flame with fluorine atoms. The flame temperature can be varied to control the degree of dissociation. This torch represents an improvement over the plasma-torch for mitigation because it provides a combination of heat sufficient to soften the silica, and fluorine atoms to etch the silica. The torch produces a smooth, Gaussian shaped pit in the fused silica, approximately 1.5mm dia. and ~2 to 4µm deep, with no apparent lip.

A single sample was tested in slab laser facility. The sample had 4 heavy damages (produced at ~45J/cm^2, 7.5ns), three of which were treated by the flame torch; one site was not treated. Of the three treated sites tested for growth, two had apparent contamination which appeared to initiate growth at 8J/cm^2, and one apparently uncontaminated site survived 1000 shots at 8J/cm^2 at 11ns. Contamination within the treated area was presumed to be carbon from dissociated CF_4.

Although only a small set of sites were tested, the flame torch shows promise to mitigate growth of heavy laser-initiated damage. Contamination by carbon deposits is also a problem with this method. The hardware is simple and inexpensive, and it could be

configured to rapidly process multiple sites on large optics. The method needs additional development, but it could be a reasonable back-up method to the CO_2 process.

Conclusions

A suite of carefully chosen characterization tools has been applied to quantitatively elucidate the morphology, densification and defects in fused silica induced by high fluence 3ω laser pulses. These structural data for high fluence initiated damage serve as an experimental baseline to explore the damage morphology and modified chemistry of optics at the NIF relevant fluences of 10-12 J/cm^2 at 3ns pulse width. Furthermore, knowledge of the high-fluence induced microstructure and defects serve a dual function of (i) providing clues to understand the laser-material interaction processes leading to damage and (ii) suggesting viable mitigation processes for damage growth in NIF optics.

We demonstrated that chemical etching of the silica surfaces to depths greater than 5 micrometers raised the damage threshold by 30% and effectively mitigated growth for 60% of the tested sites at laser fluences from 6.9 to 8.4 J/cm^2. We achieved total mitigation of damage growth for fluences as high as 12J/cm^2 by exposing the damage sites with a single pulse of a moderate power CO_2 laser. We believe this process is successful because the rapid heating melts the silica, returning the silica to the SiO_2 stoichiometry, and mending cracks., thus eliminating both conditions believed to be responsible for damage growth.

All of the methods that were tested for actual mitigation gave promising results for mitigating heavily damaged silica. Clearly the CO_2 processing gives the most convincing results. Moreover, the CO_2 laser processing method should be relatively inexpensive and straightforward to apply for mitigating sites on large optics, on a production scale. Chemical etching the surface with HF solution also exhibits promise to mitigate heavy laser-initiated damage. However, deep chemical etching may disfigure the optic out of specifications; this needs to be determined. Nevertheless, since it is potentially a relatively inexpensive method also, it should be considered as a primary backup to the CO_2 method, for mitigating surface damage on NIF silica optics.

All of the mitigation methods locally change the fused silica to leave a pit geometry that modifies the propagated waveform. It must be determined if selected pit geometries can lead to increased probability of damage to downstream optics. Modelers have been working this problem and have shown so far, that the pit geometry (Gaussian) for the CO_2 pits should not affect downstream optics when the pit diameter is less than 0.2mm, which should be achievable with further development.

Some work has been done with global application of the CO_2 laser to modify the entire silica surface, and additional experiments are planned. However, to date, such treatments have caused the surface of the silica to be highly stressed, leading in time, to surface cracks (crazing). Some progress has been made to limit the stressing of the silica surface, so this may inevitably work. But site-by-site processing is also straightforward, and it

should be developed as the primary approach. It is reasonable to envisage a system that does the initiation, identification, and CO_2 processing steps, all at the same time.

This LDRD project has provided a basic and detailed understanding of the morphology, microstructure and defect chemistry of the initiated damage sites as well as the damage growth behavior, leading us to a successful mitigation process.

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